

Attachment 2

Addendum to the Mercury Variance Application for Clean Water Services

Introduction

Clean Water Services submitted a variance application for methylmercury to the Oregon Department of Environmental Quality (ODEQ) on August 29, 2017. The variance was requested due to naturally occurring concentrations preventing attainment of the criterion and human-caused pollution sources preventing attainment of the criterion that cannot be remedied. At the request of ODEQ, supplemental data and/or literature was requested to further illustrate that the dominant sources of mercury in the Tualatin River Watershed, which prevent attainment of the criterion, are similar to those throughout Oregon and the region, and that fish tissue concentrations above the Table 40 standard of 0.04 mg/kg are endemic throughout the state and region. This addendum provides additional data analysis and cites additional literature supporting these assertions beyond what was present in the original variance application. Specifically, the data and literature cited in this addendum support the following

statements:

1. Atmospheric deposition is a dominant source of mercury to surface waters throughout the region.
2. The sources of mercury in the atmosphere, which is deposited throughout Oregon, are local, regional and global.
3. Mercury concentrations in fish tissue very frequently exceed 0.04 mg/kg across the state, region and nation, even in remote locations.
4. A portion of the mercury in fish tissue is from natural sources and may cause exceedance of 0.04 mg/kg in fish tissue even in the absence of anthropogenic sources.

These assertions are addressed separately in the following sections.

1. Atmospheric deposition is a dominant source of mercury to surface waters throughout the region

On a global, national and regional scale, there is a substantial amount of literature stating that, except in watersheds with significant mining effects or geologic enrichment, the primary pathway for mercury entering most aquatic systems is through deposition from the atmosphere (Wentz et al., 2014; Fitzgerald et al., 1998, Liu et al., 2012; Brunette & Gay, 2014; Eagle-Smith et al., 2016c; Herger, 2016).

Where geologic enrichment is not present, approximately 80 percent of terrestrial mercury originates from atmospheric inputs (Eagles-Smith et al., 2016c). Wet deposition alone typically makes up approximately 40-75 percent of the mercury input to surface waterbodies (Brunette & Gay, 2014), but dry deposition also occurs. In the 2006 Willamette Mercury TMDL, ODEQ estimated that atmospheric deposition makes up 47.7 percent of the estimated mass load of mercury to the Willamette Basin, with erosion of mercury-containing soils another 47.8 percent (ODEQ, 2006).

The evidence for atmospheric deposition as a dominant pathway includes actual measurements of mass fluxes of mercury from atmospheric deposition as well as inferences from patterns of mercury concentrations in fish tissue.

Evidence in deposition data

Since the TMDL was published in 2006, atmospheric mercury deposition data throughout the United States has continued to be collected and reported by the National Atmospheric Deposition Program Mercury Deposition Network (MDN). While mercury concentrations in precipitation are low to medium in Western Oregon (Figure 1), due to the relatively large amount of precipitation in Western Oregon, the mass load of atmospheric deposition is consistently one of the highest in the country (Figures 2-4).

Based on a low estimate mass flux of $18 \mu\text{g}/\text{m}^2/\text{year}$ for Western Oregon from these figures, approximately 550 kg of mercury per year is input into the Willamette Basin from wet aerial deposition alone. This value is similar in magnitude to the estimates used in the TMDL, which found atmospheric deposition to be a dominant source of mercury to the Willamette River Watershed.

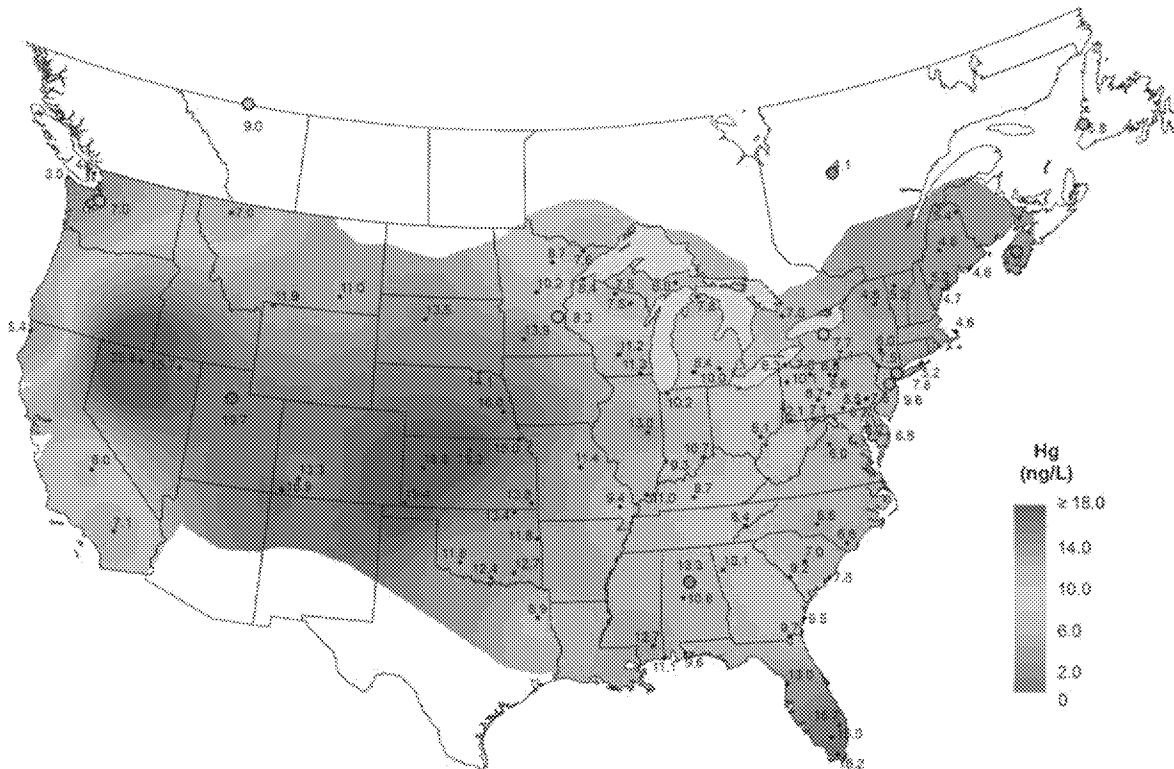


Figure 1: Precipitation-weighted mean concentration of total mercury in precipitation in 2014 (MDN, 2017)

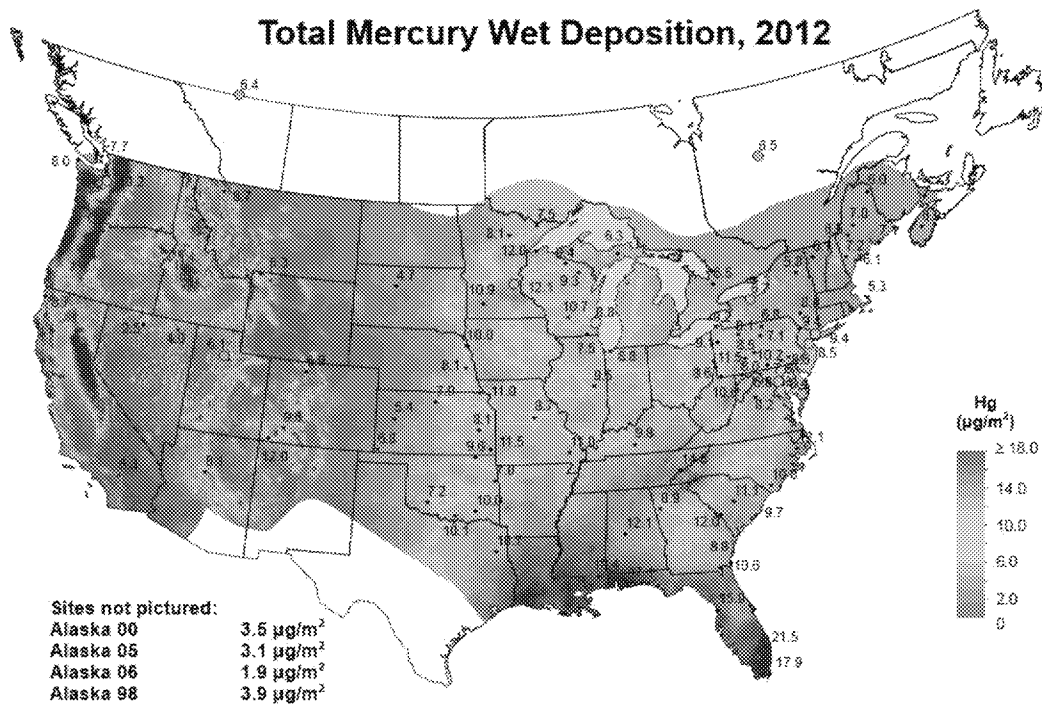


Figure 2: Total Mercury Wet Deposition in 2012 (MDN, 2017)

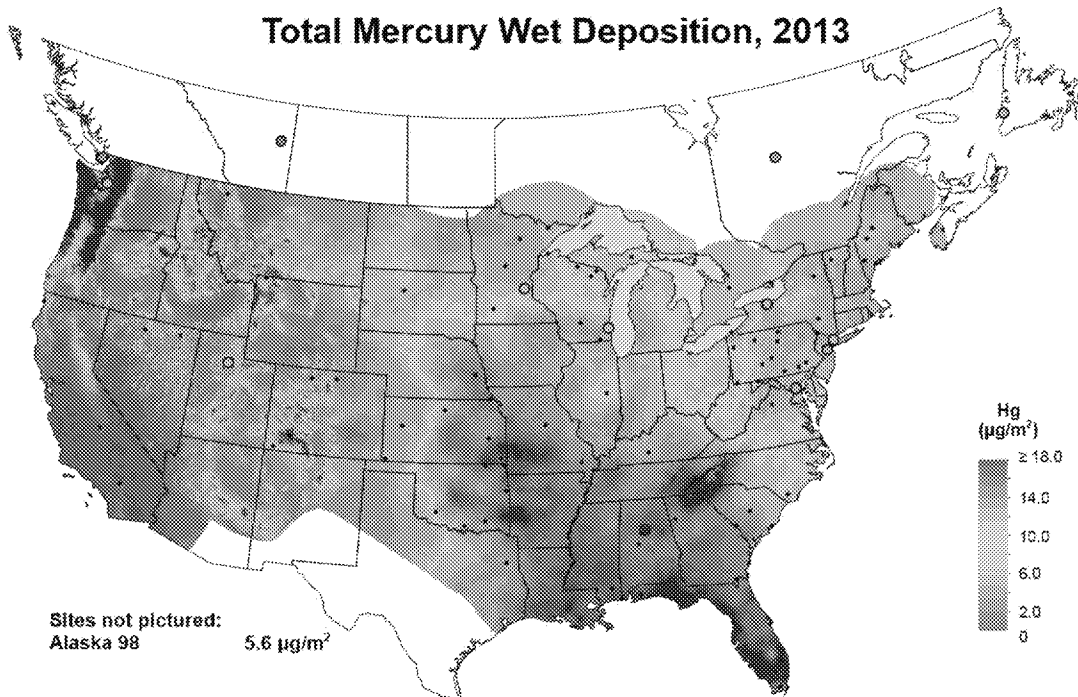


Figure 3: Total Mercury Wet Deposition in 2013 (MDN, 2017)

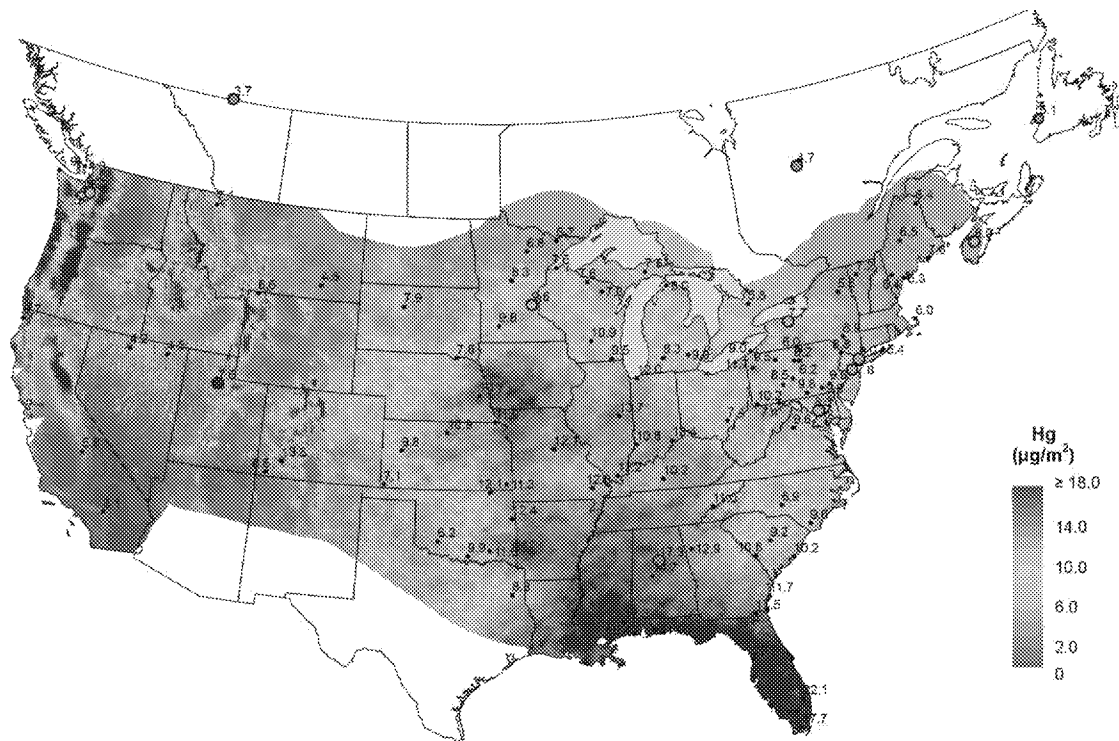


Figure 4: Total Mercury Wet Deposition in 2014 (MDN, 2017)

Evidence in fish tissue data

While analysis of atmospheric deposition data demonstrates that large masses of mercury are deposited to Oregon and the United States through atmospheric deposition, mercury concentration data in fish tissue can be used to demonstrate that it is a dominant source. Fish and other aquatic life collected in remote regions and undeveloped areas often have elevated concentrations of mercury in their tissue as has been frequently observed in Oregon (Eagles-Smith et al., 2016a; Peterson et al., 2002) and the United States (Peterson et al., 2007; Hammerschmidt & Fitzgerald, 2006). While each of the studies cited above attributed the source of elevated mercury present in fish tissue at these locations to atmospheric deposition, the presence of mercury in remote locations alone does not prove that atmospheric deposition is a dominant pathway regionally and nationally. Instead, the patterns seen within the fish tissue data from remote as well as developed locations indicate that atmospheric deposition is a dominant pathway. For example, methylmercury concentrations in fish tissues from 154 locations around Oregon were observed to be in a narrow range across a wide range of ecological conditions and habitats (Peterson et al., 2002). The author suggested that this narrow range would indicate that atmospheric transport is an important vehicle for mercury distribution throughout Oregon (Peterson et al., 2002) because a narrow range in concentrations across other variables suggests a single, dominant factor affecting wide areas. This pattern was also observed on a larger scale, when total mercury concentrations in large fish filets from 626 sites around the Western United States typically varied by less than one order of magnitude, even as other water quality attributes ranged over three orders of magnitude (Peterson et al., 2007). The fish tissue concentrations had no relationship with environmental variables such as geology or degree of anthropogenic disturbance in the watershed (Peterson et al., 2007). This uniformity suggests a widespread factor over the entire Western United

States, which strongly affected fish tissue concentrations. The authors attributed this to atmospheric deposition as “an important factor controlling regional patterns in fish tissue mercury concentrations in the Western United States” (Peterson et al., 2007).

Another line of evidence for atmospheric deposition as a dominant pathway in fish tissue data is the correlation between fish tissue mercury concentrations and wet atmospheric deposition rates. In data collected throughout the United States, methylmercury concentrations in largemouth bass filets, mosquitos and loons in most states, including Oregon, were positively correlated to wet deposition mercury flux data even while they were not correlated to mean annual precipitation, acid sulfate deposition or air temperature (Hammerschmidt & Fitzgerald, 2006). Areas with similar ranges of measured wet atmospheric deposition had similar levels of methylmercury in largemouth bass filets and loons, including Oregon, Washington and California (Hammerschmidt & Fitzgerald, 2006). The authors concluded that two thirds of the variation in methylmercury concentrations could be explained by atmospheric deposition (Hammerschmidt & Fitzgerald, 2006).

Another suggestive finding is that methylmercury concentrations in fish tissue are positively correlated with the degree of forest cover in the contributing watershed (Eagles-Smith et al., 2016a; Scudder et al., 2009). While the reasons for this may be that forests provide more organic carbon and often more wetland areas leading to more methylation, authors have also suggested this may be due to enhanced scavenging of atmospheric mercury by forests (Eagles-Smith et al., 2016a), which is a well-documented phenomenon acknowledged in the Willamette Mercury TMDL (ODEQ, 2006). The correlation of mercury concentrations in fish tissue with forest cover would, therefore, suggest runoff of atmospheric deposition as the dominant source.

2. The sources of mercury in the atmosphere, which is deposited throughout Oregon, are local, regional and global

Elemental mercury, which is emitted into the atmosphere, can travel hundreds to thousands of miles in the troposphere before being oxidized and deposited as wet or dry deposition (Liu et al., 2012; Brunette & Gay, 2014; ODEQ, 2006). The ability of mercury to persist for long periods of time and travel long distances in the atmosphere means that the mercury that is deposited in Oregon can come from local sources within the same watershed, from other locations in the state and region, from other locations in the United States and Canada and even other continents. The Willamette Mercury TMDL estimated that atmospheric deposition in the Willamette Basin was made up of 162 kg/year from local sources and 817 kg/year from global sources, meaning that approximately 85 percent of the mercury deposited was from nonlocal sources (ODEQ, 2006). This was also predicted in the mercury mass balance of the Willamette Basin that stated that local anthropogenic sources make relatively smaller contributions to the Willamette Basin than do persistent global sources (Hope, 2006).

As stated in the previous section, fish and other aquatic life collected in remote regions and undeveloped areas often have elevated concentrations of mercury in their tissue as has been frequently observed in Oregon (Eagles-Smith et al., 2016a; Peterson et al., 2002) and the United States (Peterson et al., 2007; Hammerschmidt & Fitzgerald, 2006). While weathering of geologic formations and soils containing mercury could be responsible for mercury concentrations in certain locations, geologic enrichment of mercury is rare, while the mercury in fish tissue is ubiquitous. This suggests long-range

transport of mercury and deposition in remote areas. An additional line of evidence suggesting longrange transport of mercury is increased deposition at higher elevations compared to lower elevations with similar characteristics. In the Western United States, significantly greater atmospheric deposition rates of mercury were observed at higher elevations than at lower elevations (Gustin, 2012; Wright et al., 2014). This was observed along the West Coast in Oregon and California (Gustin, 2012) as well as between the West Coast and the Great Basin between California and Nevada (Wright et al., 2014). The authors attributed the difference to mercury sources associated with long-range transport in the troposphere.

Another line of evidence is an observed spatial gradient in mercury concentrations in fish tissue decreasing from west to east both in Oregon (Peterson et al., 2002) and the United States (Wentz et al., 2014; Herger, 2016; Eagle-Smith et al., 2016c) that does not match a gradient in local sources. Across the United States, mercury discharges and atmospheric emission rates have been decreasing in recent decades, but only the Eastern United States has consistently experienced a corresponding decrease in mercury concentrations in fish tissue (Wentz et al., 2014; Herger, 2016; Eagle-Smith et al., 2016c). In each of these publications, the authors attribute this to a major source of mercury outside of the United States and Canada coming from the west, which disproportionally affects the Western United States (Wentz et al., 2014; Herger, 2016; Eagle-Smith et al., 2016c). The U.S. Geological Survey concludes that “any reductions in domestic emissions in the Western United States may be offset by increased emissions from Asia” (Wentz et al., 2014) suggesting that mercury transported in the atmosphere from Asia is a major source of mercury to the Western United States. In Oregon, mercury concentrations in fish tissue were consistently higher in the western part of the state than the eastern part of the state, which the authors suggest “may be related to atmospheric transport of Asian contaminants and rapid rain-out effect as the air mass reaches the Oregon Coast” (Peterson et al., 2002).

3. Mercury concentrations in fish tissue very frequently exceed 0.04 mg/kg across the state, region and nation, even in remote locations

The ODEQ human health water quality criteria for toxic pollutants (Table 40) lists a standard of 0.04 mg/kg of methylmercury for fish tissue in the most recent version published in 2014. This is much lower than the value assumed in the 2006 TMDL (0.3 mg/kg). There are numerous examples showing mercury concentrations in fish tissue well in excess of this standard throughout Oregon and the rest of the United States, even in remote areas removed from any local anthropogenic sources. This is especially true in large, predator and/or piscivores fish most commonly fished for by anglers. In samples collected in the late 1990s to early 2000s, piscivore fish at 154 locations throughout Oregon had an average filet mercury concentration of 0.225 mg/kg in the western region and 0.147 mg/kg in the eastern region (Peterson et al., 2002). Virtually none of the piscivores collected throughout the state had fish tissue concentrations at or below the standard of 0.04 mg/kg (equal to 0.04 µg/g) as shown in Figure 5 (Peterson et al., 2002).

ODEQ observed fish tissue concentrations across the state far in excess of 0.04 mg/kg, especially for sport fish in data collected between 2009 and 2014 (ODEQ Water Quality Toxics Monitoring Web Page, 2017; Pillsbury, 2017). The geometric mean for each species along with the range of geometric means for all sites sampled is shown in Figure 6. For most sport fish, there was not a single location that had a

geometric mean below the 0.04 mg/kg standard. For some species, there was not a single sample below the 0.04 mg/kg standard, such as Northern Pikeminnow and Largemouth Bass (Figure 7). Higher concentrations were not limited to sites with highly developed watersheds (Figure 7). In fact, in recently collected data, trout in subalpine lakes around the Pacific Northwest had a geometric mean of filet mercury concentrations of 0.043 mg/kg, which indicates that a large portion of the fish exceeded the standard where no development existed in their watershed (Eagles-Smith et al., 2016a). A 2012-2014 survey of various predator fish in 50 fishable lakes around the Pacific Northwest likewise found that 91 percent of the fish collected had mercury concentrations in their filets greater than 0.04 mg/kg (Herger, 2016). A cumulative proportion plot from Herger, 2016, illustrates that concentration ranges were similar to other studies cited above (Figure 8).

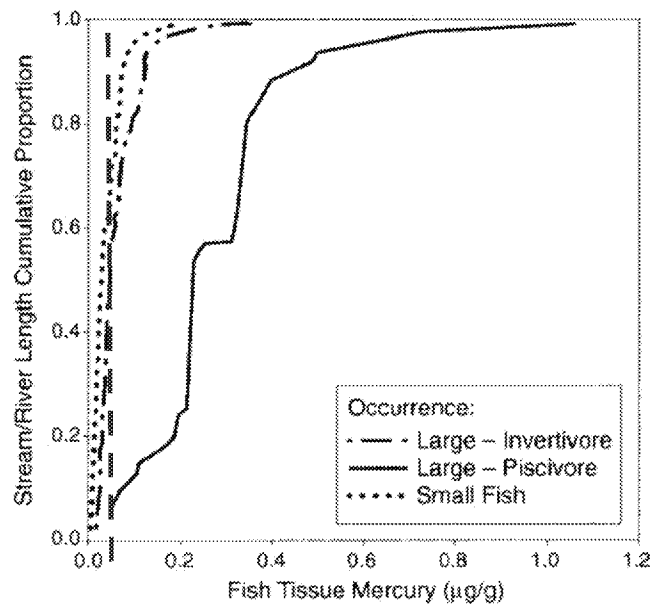


Figure 5: Excerpt from Peterson et al., 2002 (with dashed red line added for emphasis to show the 0.04 mg/kg criterion. Fish tissue mercury concentration cumulative proportion plot. Note that µg/g is equal to mg/kg.

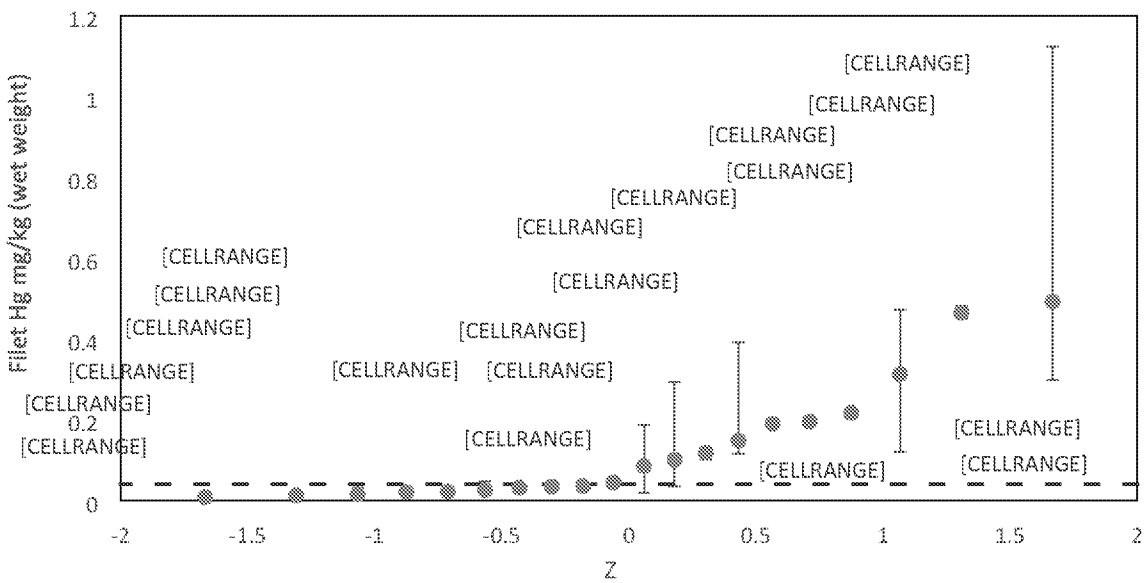


Figure 6: Geometric mean by species for all sites in data collected by ODEQ 2009-2014. Error bars represent range of geometric means for each site. Dashed red line represents the Table 40 standard of 0.04 mg/kg Hg. Z score along the axis is a measure of the number of standard deviations away from the mean.

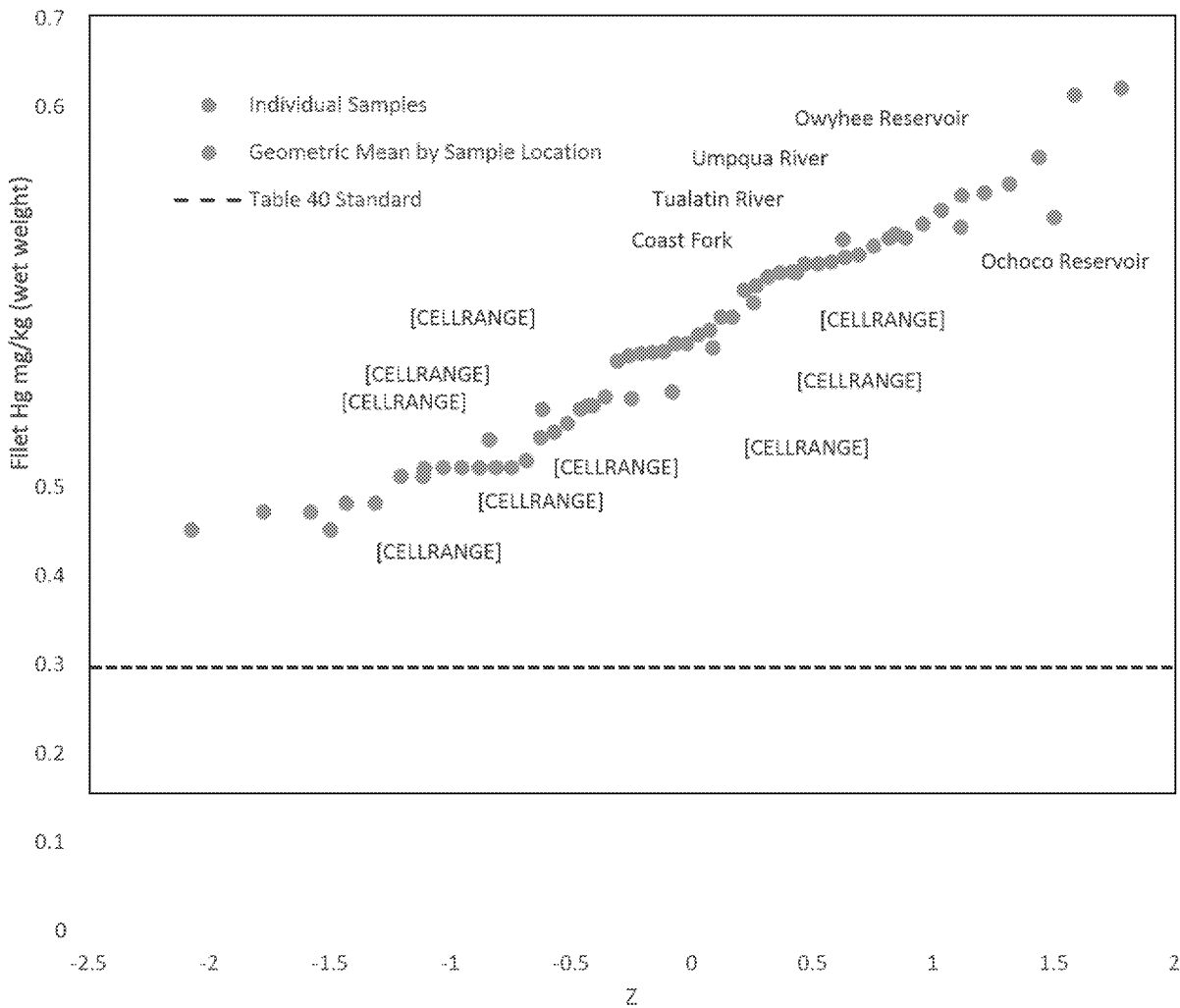


Figure 7: Mercury concentrations in smallmouth bass fish tissue in data collected by ODEQ 2009-2014. Z score along the axis is a measure of the number of standard deviations away from the mean.

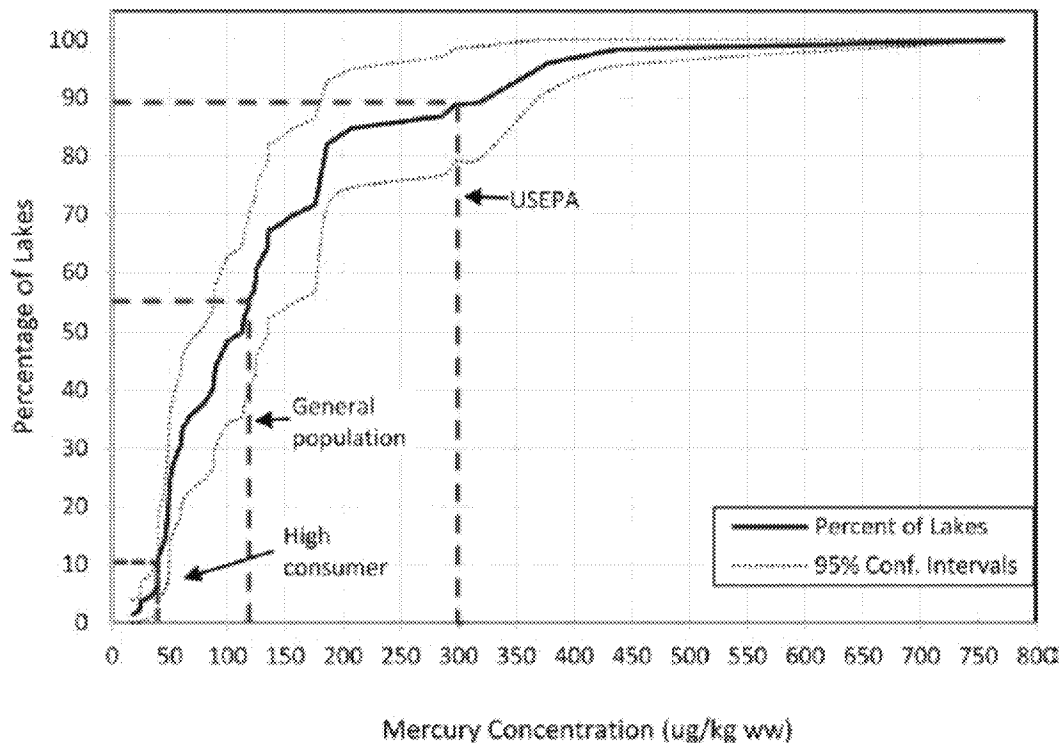


Figure 8: Cumulative proportion plot of fish file mercury concentrations from 50 lakes around the Pacific Northwest 2012-2014, excerpt from Herger, 2016. Note that the 40 $\mu\text{g/kg}$ “high consumer” value is equal to the Table 40 value of 0.04 mg/kg.

Studies of the Western United States show similar widespread presence of fish with mercury concentrations in their filets well in excess of 0.04 mg/kg. In a recent survey of mercury concentrations in fish tissue collected across various sites in Western United States and Canada, the vast majority of sites had geometric means greater the 0.05 mg/kg, as illustrated in a figure from Eagles-Smith et al., 2016b (Figure 9). No species that was sampled had a size-standardized least-squares mean lower than 0.04 mg/kg (Figure 10). The authors stated that elevated mercury contamination was widespread, including in sparsely populated areas, and that there is a complicated and diverse set of factors influencing mercury bioaccumulation across the region (Eagles-Smith et al., 2016b). A similar survey of 2,707 large fish from 626 streams and rivers across the Western United States showed that the average filet concentrations for piscivores and non-piscivores were 0.43 mg/kg and 0.14 mg/kg, respectively (Peterson et al., 2007), both far above the 0.04 mg/kg standard.

The widespread mercury concentrations in fish tissues in excess of the 0.04 mg/kg that was seen in Oregon, the Pacific Northwest and the Western United States has also been observed throughout the rest of the United States. In a survey of predator and bottom-feeding fish in 500 lakes throughout the United States, the 10th percentile and median mercury concentrations in filets from predators was 0.089 and 0.284 mg/kg, respectively (Stahl et al., 2009). Bottom-feeders also often had mercury concentrations in filets that exceeded the standard with a 10th percentile and median concentration of 0.02 and 0.069 mg/kg, respectively (Stahl et al., 2009). Therefore, less than 10 percent of predators and less than half of bottom dwellers had fish tissue concentrations that would meet the 0.04 mg/kg

standard. In largemouth bass sampled throughout the United States, state average mercury concentrations in filets ranged between 0.16 mg/kg and 0.75 mg/kg, with Oregon at 0.22 mg/kg (Hammerschmidt & Fitzgerald, 2006). A similar U.S. Geological Survey study using fish tissue data collected between 1999 and 2005 found average mercury concentrations of 0.261 mg/kg with a median of 0.126 mg/kg (Scudder et al., 2009). All of these studies reported widespread, consistent mercury concentrations in fish tissue well above 0.04 mg/kg.

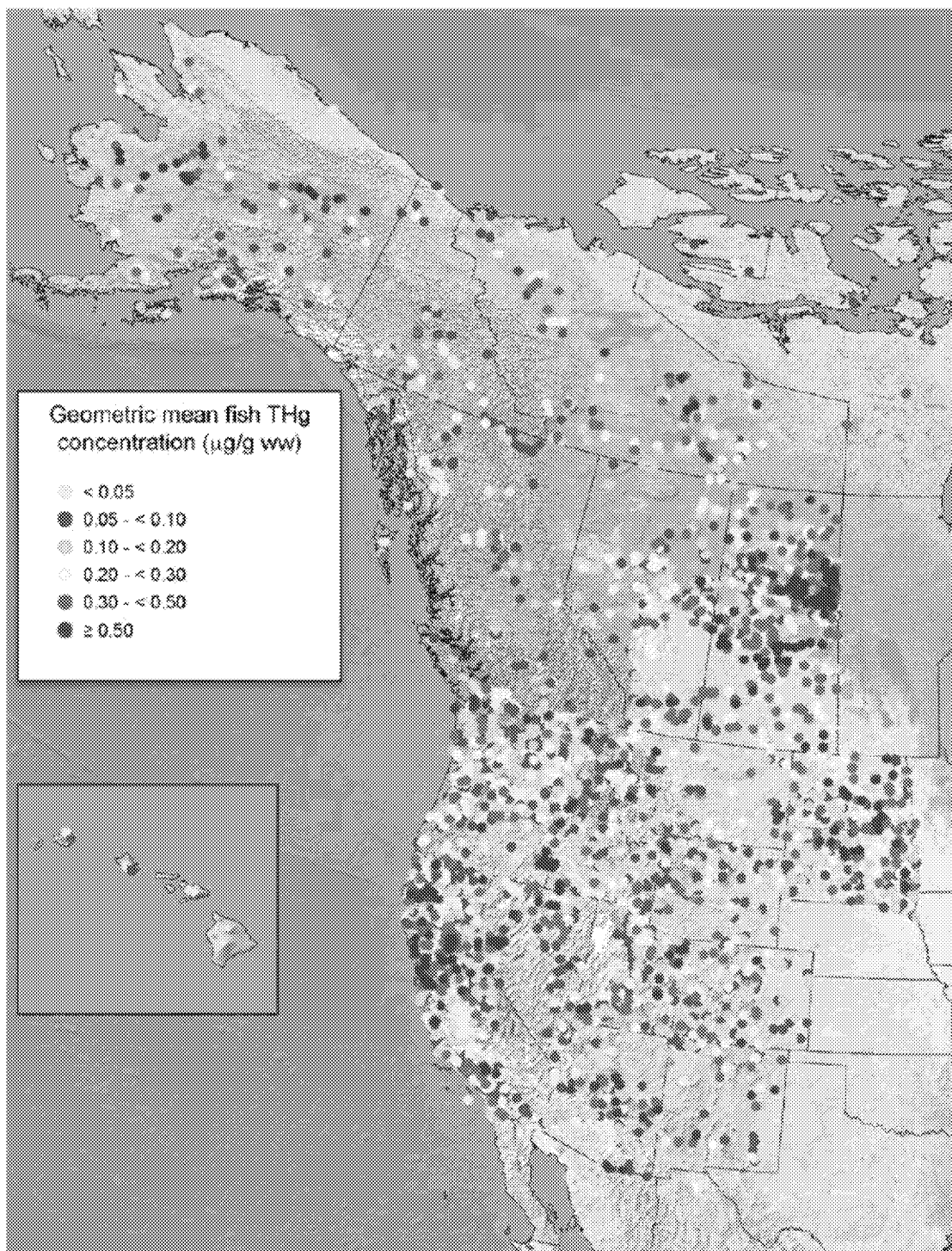


Figure 9: Figure from Eagles-Smith et al., 2016b. Geometric mean of fish tissue concentrations by site. Note that $\mu\text{g/g}$ is equal to mg/kg . Only locations with turquoise dots would have geometric means close to the 0.04 mg/kg standard.

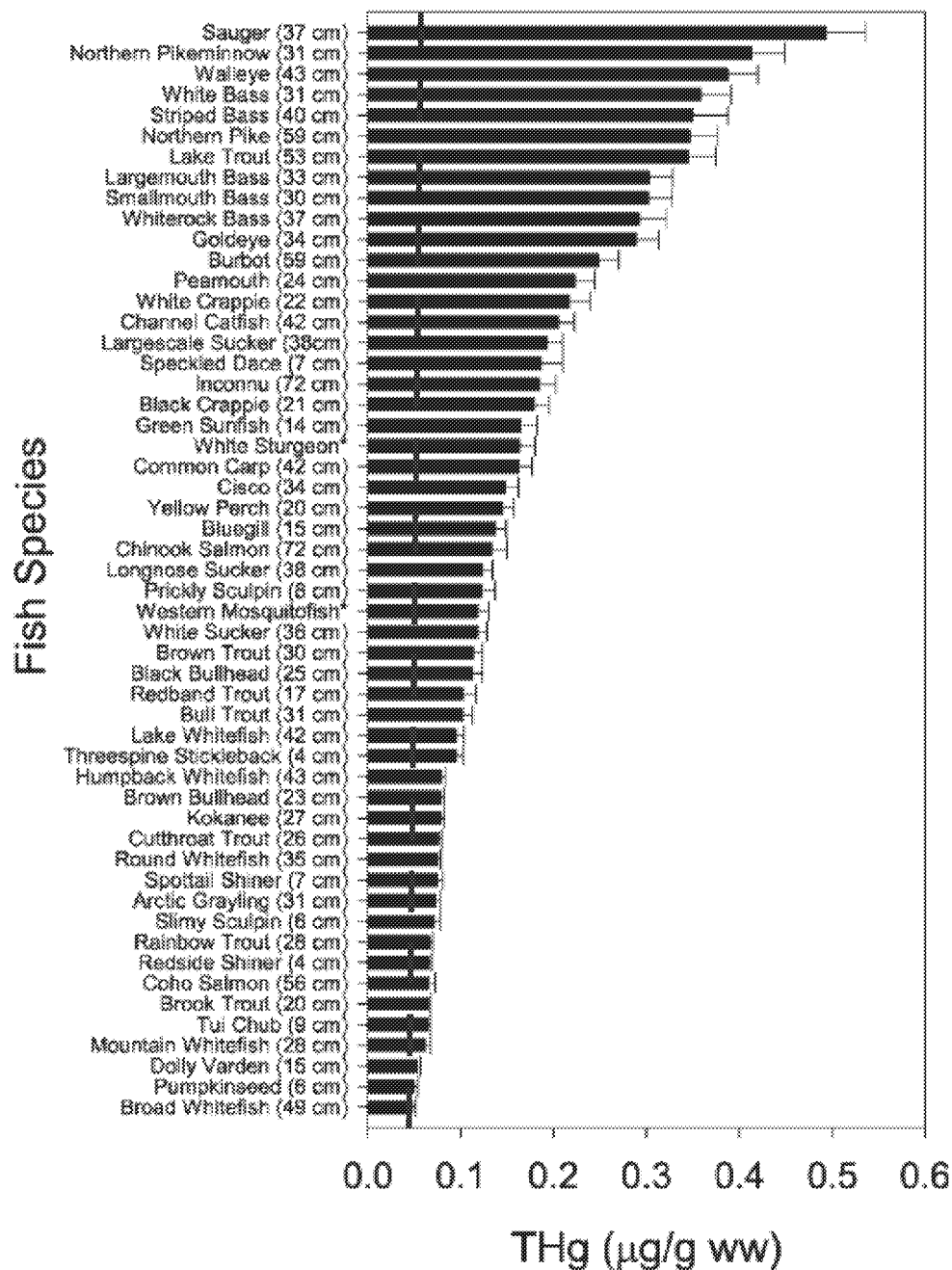


Figure 10: Figure from Eagles-Smith et al., 2016b (with red dashed line added for emphasis to represent the 0.04 mg/kg criterion). Least squares mean, size standardized muscle tissue total mercury concentrations in fish species across Western United States and Canada. Data represent species with a total samples size of >100 individuals. Error bars represent 1 standard error. Least squares mean concentrations represent the mean mercury concentration in each species after controlling for site and year effects. Fish concentrations were standardized to the respective median length for each species, shown in parentheses.

* Indicates there weren't length-mercury relationships, so the concentration is not size-standardized.

4. A portion of the mercury in fish tissue is from natural sources and may cause exceedance of 0.04 mg/kg in fish tissue even in the absence of anthropogenic sources

Mercury is introduced to the environment through natural (e.g. volcanoes, volatilization from marine environments, geothermal springs, rock weathering and forest fires) and anthropogenic sources (coal combustion, industrial waste incinerators, mining and mercury-containing products) found throughout the world (ODEQ, 2006; Herger, 2016; Brunette & Gay, 2014; Lamborg et al., 2002; Liu et al., 2012, Wentz et al., 2014). However, determining the fraction of the mercury in the environment that is from anthropogenic sources is difficult. The 2006 Willamette Mercury TMDL acknowledged that it is unclear what fraction of mercury originated from natural sources due to “paucity of literature values and sitespecific information from the Willamette Basin” (ODEQ, 2006). Some studies have suggested that anthropogenic activities have increased the atmospheric mercury concentration between two and five times since the Industrial Revolution (Liu et al., 2012; Wentz et al., 2014; Lamborg et al., 2002). While the atmospheric concentrations remains enriched to some extent, decreases in emissions in recent decades have lowered such that approximately half of all current emissions to the atmosphere are from anthropogenic sources (Liu et al., 2012, Wentz et al., 2014), approximately 80 percent of which comes from combustion (Liu et al., 2012).

Because mercury in the environment comes from both natural and anthropogenic sources, a fraction of the mercury in fish tissue is also from natural sources. It is difficult to determine what fraction comes from natural sources because mercury from anthropogenic sources is ubiquitous and mixed with natural “background” mercury throughout the globe. However, if fish tissue concentrations can be estimated from the pre-Anthropocene Era before any anthropogenic effects took place, this may approximate the natural “background” present from similar natural processes today. Fish tissue from the preAnthropocene Era is, of course, not available to be sampled. However, data from peat bogs and sediment deposition can help provide insight into pre-anthropogenic atmospheric mercury concentrations. This data has been used to estimate mercury concentrations in fish tissue from before the Industrial Revolution (USEPA, 1997) and before the Anthropocene Era (Hope & Louch, 2013) using environmental mercury cycling modeling under a range of scenarios. An excerpt from Hope & Louch, 2013 in Figure 11 shows the results from both studies. In all but one scenario (river), the estimated median predator fish tissue concentration is well above 40 µg/kg (equal to 0.04 mg/kg) for both modeling studies. While models are estimates, not measurements, the agreement of the separate modeling efforts suggests that it is likely that the natural “background” of mercury concentrations in fish tissue could very frequently exceed 0.04 mg/kg, regardless of the presence of anthropogenic sources.

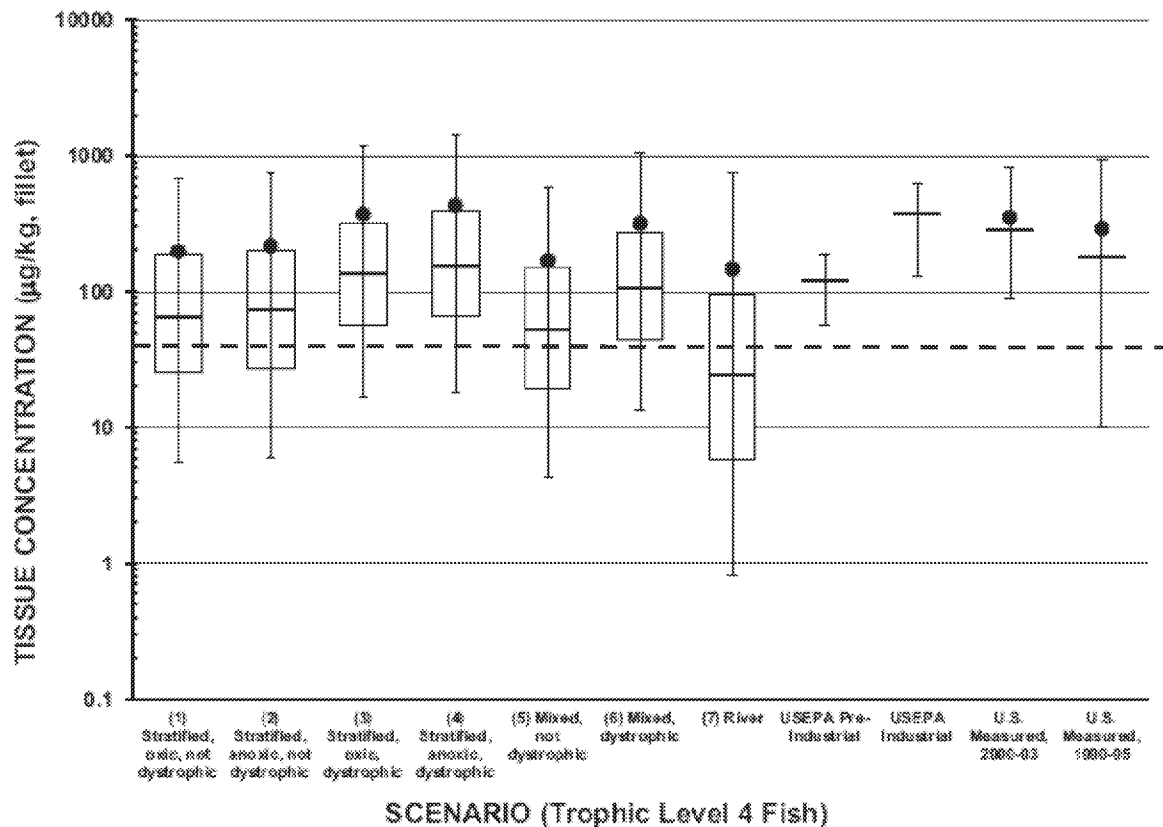


Figure 11: Excerpt from Hope & Louch, 2013 (with dashed red line added for emphasis to represent the 0.04 mg/kg criterion). Estimated and measured mercury concentrations in filets of trophic level 4 (predator) fish. (1)-(7) are model estimates of preAnthropocene concentrations given various lake and river scenarios. USEPA Pre-Industrial and USEPA Industrial are singlescenario model estimates from USEPA, 1997. Ninety-fifth and 5th percentiles are denoted by the whiskers, the 25th and 75th percentiles are denoted by the box, the median is the thick line within the box and the mean is the dot.

Conclusion

The mercury variance request submitted by Clean Water Services made the argument that humancaused pollution sources that cannot be remedied and naturally occurring concentrations prevent attainment of the criterion. The literature cited in this addendum confirms what was stated in the TMDL that atmospheric deposition is a major pathway for mercury inputs to surface waters in Oregon, the Pacific Northwest and the United States and that a large fraction of these inputs are from nonlocal sources. Controlling atmospheric emissions from local and especially non-local sources is well outside the ability or authority of Clean Water Services. Therefore, human-caused pollution sources prevent attainment of the criterion that cannot be remedied. The literature also shows that fish tissues throughout Oregon, the Pacific Northwest and the United States very frequently exceed the Table 40 criterion of 0.04 mg/kg, even in areas not impacted by point sources or nonatmospheric anthropogenic impacts. Modeling studies suggest that, while anthropogenic inputs do increase the mercury concentrations in fish tissue even without any anthropogenic inputs, it is likely that mercury concentrations in fish tissues would frequently not meet the 0.04 mg/kg criterion. Combined with the other natural sources in the watershed discussed in the original variance request, it is clear that a

combination of naturally occurring concentrations and anthropogenic sources that cannot be controlled prevent attainment of the criterion.

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